DYNAMICS OF ELECTRONS AND AB-INITIO MODELING OF QUANTUM TRANSPORT

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INTRODUCTION

In the last decade substantial attention has been given to transport of electrons through individual atomic point contacts or molecules, commonly referred to as quantum junctions (QJ). New experimental results for I(V) curves of QJ are promptly followed by their numerical modeling which, however, frequently lead to results that differ by order of magnitude from the measured data [1]. These discrepancies triggered interest in the reliability of the often quietly assumed approximations. Prime suspects are incorrect atomic geometries, inappropriate description of the exchange-correlation effects based on ground state methods and possible necessity to use a full time-dependent simulation to arrive to a correct current-carrying stead-state.

In this short paper we first give a very simple derivation of the Landauer formula for a 2-point conductance of QJ G^{2P} , based on the uncertainty principle. The aim of this is to introduce this central equation of quantum transport to a general audience. Next we analyse the dynamics of setting up a steady-state current in a simple many-electron system and use these observations to present physical basis and formal result for the 4-point conductance G^{4P} , rigorously related to the non-local conductivity of an extended system consisting of electrodes and their junction.

LANDAUER FORMULA AND THE UNCERTAINTY PRINCIPLE

Let us consider a general QJ. Single-particle quantum states - wavefunctions that can be occupied with electrons and carry current - extend with nonzero amplitude from the left electrode through the junction into the right electrode. These states, as in every metallic system, form a continuum of states of energy $E \in (E_{min}, E_{max})$, where E_{min} and E_{max} are the bottom and the top of the conduction band of the electrodes in equilibrium. In equilibrium the Fermi energy E_F is located somewhere within this interval. Applying bias voltage ΔV between the two electrodes means that those states within the continuum that carry current to the right (right-going scattering states) will be occupied up to energy μ_L , that is, $e\Delta V$ higher than the states that carry current to the left (left-going scattering states), occupied up to the energy μ_R , i.e. $e\Delta V = \mu_R - \mu_L$. Since degenerate energy levels with both right- and left- going states occupied carry zero total current, the only contribution to the transport of charge originates from the energy interval (μ_L, μ_R) , occupied only with the right-going states.

At this point we depart from the traditional derivation [2] and make use of the uncertainty principle. Right-going electrons occupying states from the energy interval (μ_L, μ_R) can be put into wavepackets with energy uncertainty $\Delta E = \mu_L - \mu_R = e\Delta V$. The uncertainty principle then states that there is an uncertainty in time Δt within which we can observe one electron passing through the junction

$$\Delta E \Delta t \sim h.$$
 (1)

However, there might be only some probability $T(E_F) < 1$, depending in general on the energy¹, that an electron will pass through the junction, out of N electrons approaching the junction within time interval $N\Delta t$ only fraction $NT(E_F)$ will actually get through. Using these observation and the fact that current is the number of electrons per time, we find

$$I = 2e \frac{N \sum_{i} T_i(E_F)}{N \Delta t} \sim \frac{2e^2}{h} \sum_{i} T_i(E_F) \Delta V \qquad (2)$$

where we have added a factor 2 to account for the spin degeneracy and sum over possibly several degenerate right-going states i. This is the celebrated 2 point Landauer formula [2] relating the current and the difference between electrochemical potentials in electrodes ΔV , the basic equation of modern mesoscopic physics. The quantum of conductance $G^0 = \frac{2e^2}{h} = (12.9k\Omega)^{-1}$ is obtained for "open" QJ with $T_i(E_F) = 1$ for only one i. Landauer equation represents an important result - it relates quantum-mechanical properties of QJ - the transmission probability - with the macroscopically measured quantity - the conductance. We would like to note that the weak "proportional to" sign can be made into strong "equal" introducing occupationadapted orthogonal wavepacket basis set. Including the mean-field, local-neutrality arguments leads to the 4point conductance, e.g. in 1D $G^{4P} = G^{2P}/(1-T(E_F))$ relating the current to the induced electrostatic drop in potential in the vicinity of QJ, ΔV^i [2].

DYNAMICS OF 1D QUANTUM GAS OF ELECTRONS

While previous derivation gives some hint of the time-dynamics in quantum transport, namely that based on wavepackets of electrons having significant amplitude in a given space for certain time Δt , the approach is really describing a steady state. So we ask - how do we make a transition from equilibrium to steady non-equilibrium situation in QJ?

Consider a 1D electron gas of density n, fixed by the Fermi energy $E_F = \frac{\hbar^2 k_F^2}{2m}$. At time t=0 we apply a localized electric field of the form

$$E^{e} = -\frac{\Delta V}{a} (\theta(x + a/2) - \theta(x - a/2)),$$
 (3)

where $\theta()$ is the unit step function, a the distance on which the field is nonzero, modeling the width of QJ,

Assuming smooth dependence of the transmission probability on the energy and that the applied bias is small we set $T(E) \simeq T(E_T)$ for $E \subseteq (u_T, u_T)$

and ΔV corresponds to the applied voltage. The response of the density and the current can be found using the linear response theory [3]. In general, the non-local conductivity $\vec{\sigma}$ gives a causal linear relationship between the total electric field and the current density

$$\vec{j}(\vec{r},t) = \int_0^t dt' \int d^3r' \vec{\sigma}(\vec{r}, \vec{r}'; t - t') \cdot \vec{E}(\vec{r}', t')$$
 (4)

For our simple system the nonlocal conductivity, calculated directly from the continuum of occupied quantum states, is known analytically [3]. Using this within the general formula (4) and performing a simple numerical calculation we find that the current is indeed settling to a steady value $I = \frac{2e^2}{h}\Delta V$ with a well defined relaxation time τ (see inset in Fig.1). It is instructive to analyze the dependence of the latter on the width of the junction a, while keeping the overall bias voltage ΔV constant. The resulting dependence for a gas with $E_F = 0.07 \mathrm{Ha} = 2 \mathrm{eV}$ (corresponding to a gold nanowire) is shown in the Fig.1. The behavior of τ has a nice physical interpretation: for junctions

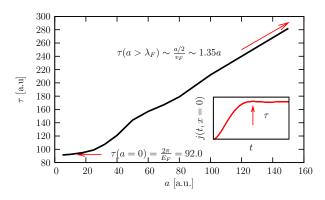


Fig. 1. Dependence of the relaxation time τ (defined by the first local maximum in j(t), see the inset) on the width of the junction a. The arrows show the limiting behaviour.

smaller than the Fermi wavelength λ_F , the relaxation time is constant and given by the timescale dictated by the Fermi energy, for junctions larger than Fermi wavelength the relaxation is related to a time it takes for an electron with Fermi speed to pass the region of the junction. These limiting results are obtainable by a detailed analysis of the analytical formulas as well.

These results demonstrate the mechanism how the current is established for non-interacting electrodes, which is pertinent to all present ab-initio calculations. However, as soon as we include even Hartree interaction, the process changes dramatically. Namely, the observed current in Fig.1 leads to charge imbalance there are electrons piling up on the right of the junction and electrons being depleted on the left of the junction. Physically this is almost obvious as any localised electric field should be screened out by electrons. The screening can be overcome only in the limit $a \to \infty$, $E^e \to 0$, $\Delta V = const$ when the field becomes homogeneous. However, as the Fig. 1 clearly shows, in this case the relaxation time becomes infinitely as well!

The resolution is following. To set a steady current

on a external homogeneous field of finite magnitude for a finite time $t < t_e$. This is to be compared with the infinitesimal magnitude and for all positive time in the previous paragraph. In the semi-infinite electrode this will create an uniform current even for $t > t_e$ and there is no problem with charge accumulation. Only in the region of QJ the charge will pile up on the left and deplete on the right. This will lead to a induced field E^i , localised at the QJ and characterised with some induced drop in potential ΔV^i . This induced field will self-consistently develop to such a form that the current I, established throughout the semi-infinite electrodes, will be able to pass the junction.

GENERAL EXPRESSION FOR CONDUCTANCE

The above ideas can be nicely formalised within the framework of the linear response theory. The current is given by

$$I(x,t) = \int_0^t dt' dx' \sigma(x, x'; t - t') \left(E^e(x', t') + E^i(x', t') \right).$$
(5)

Dividing the conductivity into the electrode's, translationally invariant part and the part primarily due to the junction, $\sigma(x, x') = \sigma^H(x - x') + \sigma^J(x, x')$ respectively, and performing the long-time analysis we arrive at the final expression for the steady-state current [4]

$$I = -\frac{\alpha}{\beta} G^{2P} \Delta V^i = G^{4P} \Delta V^i, \tag{6}$$

where

$$\alpha = \sigma^H(q = 0, t \to \infty), \tag{7}$$

$$\beta = -\int dq \sigma^J(q, q' = 0; t \to \infty),$$
 (8)

$$G^{2P} = \int \frac{dqdq'}{2\pi} \sigma(q, q'; t \to \infty), \tag{9}$$

where q, q' are wavenumbers, reciprocal to x, x' respectively. The equation (6) is expressing the 4-point conductance in terms of the limiting character of the non-local microscopic conductivity. The utility of this formulation lies in a formally straightforward evaluation of the latter using various ab-initio and, unlike the Eq. (2), also correlated many-body methods.

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